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PATENT

Docket No.: 15275/8610 (Dobbins 2-1)

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Reissue Application No.: 08/833,620)
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Filed : April 7, 1997) Examiner: J. Hoffman
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U.S. Patent No. : 5,043,002) Group Art Unit: 1731
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Granted : August 27, 1991)
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Patentees : Michael S. Dobbins and Robert E. McLay)
)
For : METHOD OF MAKING FUSED)
SILICA BY DECOMPOSING)
SILOXANES)
)

DECLARATION OF MICHAEL S. DOBBINS UNDER 37 CFR § 1.132

I, MICHAEL S. DOBBINS, pursuant to 37 CFR § 1.132, declare:

1. I received a B.S. in Chemical Engineering in 1978 from San Jose State University as well an M.S. and Ph.D. in Chemical Engineering in 1982 and 1986, respectively, from Iowa State University.
2. I am currently employed by assignee Corning, Inc. and hold the position of Business Technology Manager.
3. I am a co-inventor of the above patent application.
4. I am familiar with European Patent No. 38,900 to Schwarz et. al. ("Schwarz") which relates to a method of manufacturing a silicic acid dispersion having a specific BET surface of 250 ± 25 to 350 ± 25 m²/g and a thickening viscosity of 4000 to 8000 mPas in unsaturated polyesters by burning a polycyclosiloxane (e.g., hexamethylcyclotrisiloxane or octamethylcyclotetrasiloxane) or mixture of siloxanes in the presence of hydrogen or hydrocarbon.
5. At the time I made my above invention, researchers in this field would not have expected the polycyclosiloxanes of Schwarz to be useful in producing a non-porous body of high purity fused silica glass. In particular, Schwarz's polycyclosiloxanes have a

large number of carbon atoms and not all of these carbon atoms would have been expected to combust when the siloxanes are passed through a burner. Indeed, the production of carbon when burning polycyclosiloxanes to form pyrogenic or fume silica or silicic acid is well documented in the literature. GB 2,049,641 to Kratel et. al., (page 1, lines 6 to 28) states the following:

Very finely divided silica (highly disperse silica) may be manufactured by flame hydrolysis, which comprises reacting a gaseous or vaporisable silicon compound and, optionally, another gas that will burn to form water, with oxygen in a flame (see, for example, DE 900 339, U.S. 2,399,687, G.B. 17325/77 Serial No. 1562966 (equivalent to DE 26 20 737 A1)). Silica manufactured in this manner is known as pyrogenic silica or fume silica. Satisfactory results can be obtained by this method when silicon tetrachloride is used as the gaseous silicon compound. It is, however, often advantageous to use an organosilane as the gaseous silicon compound, but the silica produced from these compounds tends to be contaminated with carbon and thus tends to be dark in colour. This result when using silicon compounds containing silicon-bonded organic groups, especially halogen-containing silicon compounds, has previously been counteracted by using an additional fuel, namely an additional gas that will burn to form water, for example hydrogen or hydrocarbon.

In addition, J. Lipowitz, "Flammability of Poly(Dimethylsiloxanes). 1. A Model for Combustion," J. Fire & Flammability 7: 482-503 (1976) states:

Under fuel-rich conditions ($\phi > 1$), two-stage combustion is evident by formation of inner and outer flame cones. The outer flame cone is a pale bluish color typical of the outer cone of organic flames, both representing further combustion of CO and H₂. However, the inner cone is blue at $\phi < 2.67$ and luminous yellow-white at $\phi > 2.67$. Organic flames are luminous yellow due to blackbody radiation from carbon particles. Significantly, adiabatic calculations show carbon formation from D₄ and MM at $\phi > 2.67$.

* * *

Gray-brown amorphous silica collected above diffusion flames, which are luminous (yellow-white), contains 3-4% total C, primarily elemental carbon, and 0.1-0.2% H. A weak infrared band indicating some C-H bond structure is present at 2925 cm^{-1} . No crystallinity indicative of $\text{SiO}_2(\text{g})$, graphite, SiC, or Si_3N_4 is found by x-ray powder diffraction or electron diffraction. Detectability limits are estimated at several %. Particulates consist of 100\AA diameter particles tightly aggregated into $700\text{-}1000\text{\AA}$ clumps which are further agglomerated in chains (Figure 5). Some larger particles ($2000\text{-}4000\text{\AA}$ diameter) appear to be carbon and should contribute appreciably to the observed luminosity.

6. When, in accordance with the teachings of Schwarz, the dispersion product is used as a thickener, the existence of carbon in the dispersion would not present any difficulties. In fact, the presence of carbon would most likely have improved thickening.

7. However, the presence of carbon impurities could result in significant problems when making a non-porous body of high purity fused silica glass, particularly where that glass is used to make precision optical products like optical waveguide fibers. In making a non-porous body of high purity fused silica glass, scientists skilled in this area would not want any carbon to be present, not even at a parts per million level. Their concern at the time I made my invention would have been that production of carbon during glass formation could adversely affect light transmission in a number of ways. In particular, the presence of carbon particles in the glass would absorb light and cause light scattering resulting in transmission losses. Moreover, carbon particles in the glass would also be likely to associate with adjacent oxygen atoms that otherwise form silicon dioxide, resulting in C-O bond formation within the glass. Such C-O bonds would absorb light in the infrared region of the spectrum and cause transmission losses which prevent the light from traveling as far. This is a particularly significant problem in optical fibers where transmission is in the infrared region of the spectrum and such light must travel long distances. Carbon formation would also have been expected to physically remove oxygen (otherwise in the form of silicon dioxide within the glass) away from the glass as carbon monoxide. This would result in an electron deficiency in the resulting glass that would cause transmission loss in the ultraviolet

region of the spectrum. Thus, the formation of carbon in a non-porous body of high purity fused silica glass, such as that used in optical fibers or precision lenses, would be highly undesirable. In view of the recognition in the art that passing polycyclosiloxanes through the flame of a burner would have been expected to produce carbon, scientists making a non-porous body of high purity fused silica glass would not have wanted to make such products by burning polycyclosiloxanes.

8. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: 22-Nov-99



Michael S. Dobbins